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Estimating Mercury Exposure of Piscivorous Birds and Sport Fish Using Prey Fish Monitoring

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Supporting Information

ABSTRACT: Methylmercury is a global pollutant of aquatic ecosystems, and monitoring programs need tools to predict mercury exposure of wildlife. We developed equations to estimate methylmercury exposure of piscivorous birds and sport fish using mercury concentrations in prey fish. We collected original data on western grebes (*Aechmophorus occidentalis*) and Clark's grebes (*Aechmophorus clarkii*) and summarized the published literature to generate predictive equations specific to grebes and a general equation for piscivorous birds. We measured mercury concentrations in 354 grebes (blood averaged 1.06 ± 0.08 μg/g ww), 101 grebe eggs, 230 sport fish (predominantly largemouth bass and rainbow trout), and 505 prey fish (14 species) at 25 lakes throughout California. Mercury concentrations in grebe blood, grebe eggs, and sport fish were strongly related to mercury concentrations in prey fish among lakes. Each 1.0 μg/g dw (~0.24 μg/g ww) increase in prey fish resulted in an increase in mercury concentrations of 103% in grebe blood, 92% in grebe eggs, and 116% in sport fish. We also found strong correlations between mercury concentrations in grebes and sport fish among lakes. Our results indicate that prey fish monitoring can be used to estimate mercury exposure of piscivorous birds and sport fish when wildlife cannot be directly sampled.

INTRODUCTION

Methylmercury is a globally pervasive pollutant that is toxic to fish and wildlife and biomagnifies primarily through aquatic food chains.[1,2] Continued anthropogenic mercury emissions over time[2] and documented effects on fish and wildlife[3] have led to expanded efforts to monitor mercury in the environment.[4,5] Piscivorous birds are top predators in many aquatic habitats, making them among the most vulnerable taxa to methylmercury exposure and associated adverse effects.[3] However, environmental mercury monitoring programs assessing contamination of aquatic biota typically sample fish rather than wildlife due to their relative ease of sampling and often more direct link to human exposure.[5−7] As a result, the risk of methylmercury exposure to aquatic wildlife often is not known in many mercury monitoring programs.

Direct measurement of mercury concentrations in aquatic wildlife is the preferred approach for assessing their methylmercury exposure. Yet, sampling aquatic wildlife, such as piscivorous bird blood[8] and eggs,[9] often is logistically less feasible, and data associated with mercury contamination of biota are far more prevalent for fish than for wildlife. For example, in a recent synthesis of mercury data for the Great Lakes region,[10] assessments of methylmercury exposure were based on sample sizes of >43 000[11] and >63 000[12] sport fish and >6000 prey fish,[6] compared to <2000 birds.[13] Therefore, the ability to estimate mercury exposure of piscivorous birds based upon mercury concentrations in fish would be advantageous for many mercury monitoring programs. In particular, small fish often are prey for larger fish and wildlife, so they could serve as a useful indicator of methylmercury exposure to higher trophic level animals. Several large contaminant monitoring programs have incorporated prey fish sampling[13,14] under the assumption that contaminant concentrations in prey fish are a reliable indicator of risk to fish-eating wildlife. However, correlational models between mercury concentra-

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tions in piscivorous birds and prey fish are available only for common loons (*Gavia immer*)[13,15,16] and do not exist for other species of birds. Thus, it is important to assess the strength and validity of these relationships in areas where other bird species are the dominant piscivores.

Herein, we conducted a detailed assessment of the validity of using mercury concentrations in prey fish to predict mercury concentrations in piscivorous birds and sport fish. Specifically, we developed predictive equations to link mercury concentrations in small prey fish to piscivorous birds and sport fish so that mercury monitoring programs that sample fish, and do not directly sample wildlife, can more easily estimate risk of methylmercury exposure to piscivorous birds. We collected original data on mercury concentrations in western grebes (*Aechmophorus occidentalis*), Clark's grebes (*A. clarkii*), sport fish, and prey fish, and summarized the published literature on piscivorous birds to generate predictive equations specific to grebes as well as a general equation for piscivorous birds.

EXPERIMENTAL SECTION

Grebe Sampling. We sampled grebes and fish at 25 lakes and reservoirs (hereafter termed lakes) throughout California from April through October of 2012 (13 lakes) and 2013 (12 lakes; see Figure S1 of the Supporting Information, SI). We captured an average of 14 grebes per lake (range: 2−38 grebes) with night-lighting techniques[17,18] and recorded standard data on bird morphometry and molt status (see SI). We then collected whole blood (≤3.0 mL) from each bird via the brachial or jugular vein with heparinized 23−26-gauge needles and a syringe. Whole blood was immediately transferred to polypropylene cryovials, held on wet ice, and then transferred to a liquid nitrogen dewar within 6 h of collection. Blood was then transferred to the laboratory for storage at −20 °C until mercury analysis. We also collected a drop of blood from each grebe for sex determination through genetic analysis (Zoogen Services, Davis, California, U.S.A.).

At 7 of the 25 lakes where we sampled adult grebe blood, we also collected on average 14 grebe eggs (range: 6−23 eggs). We randomly collected one egg from each nest, and classified it as either randomly sampled from an active nest (random egg) or salvaged from a nest that had been abandoned before our visit (abandoned egg). We floated eggs to determine embryo age[19] and estimated nest initiation date by subtracting the clutch size and embryo age from the date the nest was visited.

We stored eggs on wet ice in the field and transferred them to a refrigerator until dissection. During egg dissection, we measured the length and width of each egg to the nearest 0.01 mm with digital calipers (Fowler, Newton, Massachusetts, U.S.A.) and measured the total egg weight to the nearest 0.01 g on a digital balance (Ohaus Adventurer Pro, Ohaus Corporation, Pine Brook, New Jersey, U.S.A.). We then cut an approximately 20 mm diameter hole in the top (air-cell end) of each egg using clean, stainless steel scissors and removed the embryo and any remaining contents into a sterile 125 mL jar with stainless steel forceps. We then stored the egg contents at −20 °C until mercury determination.

Fish Sampling. Within an average of 11 days from the date of grebe blood sampling, we returned to each of the 25 lakes to sample prey fish and sport fish. Fish were captured via electrofishing boat (Smith-Root, Vancouver, Washington, U.S.A.) and dip nets. We collected small fish (mean: 58 mm standard length, range: 18−123 mm), which were in the size range that grebes commonly consume.[20] Efforts were made to sample the same species across all lakes; when this was not possible, we sampled fish that overlapped in trophic guild. We sampled 10 individuals from each of two prey fish species from each lake, for a total of 20 prey fish per lake, with four exceptions: at three lakes we sampled 25 prey fish (10 each of two species and 5 of a third species at two lakes; and 10, 8, and 7 for each of three species at one lake) and at one lake we sampled only 10 prey fish of a single species. In total, we sampled 505 prey fish of 14 species from 25 lakes (see Table S1). For sport fish, we collected the most common species at each lake (mean: 397 mm total length, range: 178−726 mm), which is within the size range commonly consumed by humans. We sampled 10 individuals from the most common sport fish species in each lake, with three exceptions: at two lakes, we sampled only 8 sport fish and at one lake we were only able to collect 4 sport fish, of 2 different species. In total, we sampled 230 sport fish of 5 species from 24 lakes (see Table S2). We stored fish on wet ice in the field until processing. During fish processing, we weighed each fish with a digital balance (prey fish: Smart Weigh Pro Pocket Scale, Smart Weigh, Nanuet, New York, U.S.A.; sport fish: Angyo Portable Electronic Scale, Angyo, China) and measured standard length (prey fish) or total length (sport fish) with a fish board. Thereafter, fish were stored on dry ice until they were transferred to a freezer (−20 °C), where they were stored until mercury determination.

Mercury Determination. We used total mercury (THg) concentrations as an index of methylmercury (MeHg) concentrations, because most of the Hg in fish and birds is in the more toxic MeHg form.[1,9] We determined THg concentrations in whole blood, egg contents (without the eggshell), whole-body prey fish, and skinless muscle fillets of sport fish following Environmental Protection Agency Method 7473.[21] For bird blood, we determined THg concentrations on a wet-weight basis. We thawed blood to room temperature, then homogenized it in a vortexer before weighing the blood for THg determination. For bird eggs, we dried the entire egg contents at 50 °C for 48−72 h until completely dried, reweighed egg contents to determine moisture content, and then homogenized the dried egg contents to a powder in a grinder with stainless steel blades. For prey fish, whole fish were washed in deionized water to remove any debris from the fish surface, dried at 50 °C for approximately 48 h until completely dried, reweighed to determine moisture content, and then homogenized to a fine powder with a porcelain mortar and pestle. For sport fish, we filleted the fish and used a small aliquot of muscle to determine THg concentrations on a wet-weight basis. See SI for further THg determination methods and quality assurance measures.

We report THg concentrations on a dry-weight (dw) basis for prey fish and sport fish, on a wet-weight (ww) basis for bird blood, and on a fresh wet-weight basis (fww) for eggs. THg concentrations in sport fish were estimated on a dry-weight basis using individual-specific moisture content values and wet-weight THg concentrations. THg concentrations in eggs were estimated on a fresh wet-weight basis using individual-specific moisture content of egg contents and egg morphometrics following Ackerman et al.[9] Moisture content (mean ± SE) was 75.9% ± 0.14% in bird blood (2013 only; n = 149), 75.5% ± 0.14% in bird eggs (n = 101), 75.8% ± 0.11% in prey fish (n = 505), and 78.3% ± 0.15% in sport fish (n = 230).

Statistical Analysis: Mercury by Lake. In the first stage of our analyses, we used linear mixed-effect models to estimate least squares mean THg concentrations in grebe blood, grebe

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eggs, prey fish, and sport fish for each lake. For the grebe blood model, $\log_e$-transformed THg concentration (μg/g ww) was the dependent variable, species (western grebe or Clark's grebe) and sex (male or female) were fixed effects, and lake was a random effect. For the grebe egg model, $\log_e$-transformed THg concentration (μg/g fww) was the dependent variable, species (western grebe, Clark's grebe, or unknown) and egg collection type (random or abandoned) were fixed effects, and lake was a random effect. For the prey fish model, $\log_e$-transformed THg concentrations (μg/g dw) was the dependent variable; species (Table S1), standard length, and species × length interaction were fixed effects; and lake was a random effect. The sport fish model was similar to the prey fish model, except that total length was used instead of standard length. For all four response variables, least squares means were estimated for each lake from the mixed-effect models using Best Linear Unbiased Predictors in JMP software (version 11.2.0; SAS Institute Inc., Cary, North Carolina, U.S.A.). The least squares mean THg concentrations in prey fish for each lake were then used as a covariate in the next analyses describing factors influencing THg concentrations in grebe blood, grebe eggs, and sport fish.

Statistical Analysis: Factors Influencing Mercury in Grebes and Sport Fish. In the next stage of our analyses, we used linear mixed-effect models to examine which variables influenced THg concentrations in grebe blood, grebe eggs, and sport fish. For each of these tissues, we built a set of candidate models based on potential predictor variables describing the (1) specific tissue, (2) lake attributes, and (3) THg concentrations in prey fish. For each of the three tissue types, the model structure was similar except for the variables describing the specific tissue.

For grebe blood, tissue-specific predictor variables included species (western grebe or Clark's grebe), sex (male or female), bird mass, body condition index, linear (wing molt) and quadratic (wing molt^2) terms for wing molt score, and linear (date) and quadratic (date^2) terms for sampling date. We did not allow bird mass and body condition to occur in the same model. The body condition index was estimated as an individual's residual mass divided by its mass, where an individual's residual mass was calculated as the residual from a linear regression model of bird mass and structural body size (see SI for additional details). Wing molt was calculated as the mean value of molt classification for each of the 10 primary feathers. Finally, date was standardized as the difference between the day of year the bird was captured and the median day of year for all captured birds (median day of year was 181).

For grebe eggs, tissue-specific predictor variables included species (western grebe, Clark's grebe, or unknown *Aechmophorus* grebe), egg collection type (random or abandoned), date, and date.[2] Again, date was standardized as the difference between the day of year the nest was initiated and the median day of year for all nests initiated (median day of year was 211).

For sport fish, tissue-specific predictor variables included species (Table S2), total length, and species × total length interaction. Date was standardized as the difference between day of year the sport fish were captured and the median day of year for all captured sport fish (median day of year was 204).

For each of the three tissue types, the candidate model set included several lake-specific variables, including lake area, lake perimeter, lake shape index, and elevation. The lake shape index was calculated as lake perimeter divided by the square root of lake area multiplied by 0.25.[22]

Lastly, we evaluated the influence of both lake-specific least squares mean $\log_e$-transformed THg concentrations in prey fish and lake-specific geometric mean $\log_e$-transformed THg concentrations in prey fish on THg concentrations in grebe blood, grebe eggs, and sport fish, by including them in the full candidate model set with the rule that both least squares mean and geometric mean THg concentrations in prey fish could not be included in the same model. For each of the three tissue types, models including least squares mean THg concentrations in prey fish, which statistically accounted for prey fish length and species, performed substantially better than models including geometric mean THg concentrations in prey fish. The best model that included least squares mean THg concentrations in prey fish was 5.9, 6.8, and 30.3 times more likely than the best model that included geometric mean THg concentrations in prey fish for grebe blood, grebe eggs, and sport fish, respectively (all $\Delta AICc > 3.55$). Therefore, geometric mean THg concentration in prey fish was removed as a potential variable in the final candidate model sets.

For each of the three tissue types, our final candidate model set included all additive combinations of variables (with exceptions previously noted), and a null model (a total of 3456 models for grebe blood, 384 for grebe eggs, and 480 for sport fish). In each model, $\log_e$-transformed THg concentration was the dependent variable, and lake was included as a random effect. We evaluated models using second-order Akaike Information Criterion (AICc; model with the smallest AICc was considered the most parsimonious),[23] including AICc differences between the best model and each of the other candidate models ($\Delta AICc$), Akaike weights (w_i; weight of evidence of the selected model), evidence ratios (relative weight of support between models), and adjusted relative variable importance (log-odds ratio of the posterior $[P]$ and prior $[P_0]$ variable's weights summed across all models that incorporated the variable: $\ln [(P/(1-P))/(P_0/(1-P_0))]$) as described in the SI. Adjusted relative variable importance values >0 had posterior weights that were greater than was expected by their prior weighting and were considered to be important, and values <0 had posterior weights that were less than was expected by their prior weighting and were considered to be unimportant. For brevity, we present only the set of best models that were within $\Delta AICc \leq 2$ (those considered for biological importance), the null model, and each model that was similar to the best model except one of the variables in the best model was removed (see Tables S3–S5). When examining effects of a specific variable, we estimated conditional model-averaged coefficients by only model-averaging across models where the variable was present, to better reflect the true relationship of THg concentrations with that variable. However, all other results were based on model-averaged predictions and standard errors from the full candidate model set. We report back-transformed least squares means and estimated standard errors using the delta method.[24]

RESULTS

Mercury Concentrations among Lakes. We captured 354 grebes at 25 lakes; 71% were western grebes and 29% were Clark's grebes, and 48% were female and 52% were male. THg concentrations in grebe blood averaged 1.06 ± 0.08 μg/g ww but differed between species ($F_{1,331.3} = 13.35$, $p < 0.001$) and sexes ($F_{1,328.5} = 12.58$, $p < 0.001$). Least squares mean THg concentrations in grebe blood ranged from 0.16 ± 0.02 μg/g

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ww at Big Lake to 5.16 ± 0.61 μg/g ww at Lake Berryessa (see Figure S2a).

We collected 101 grebe eggs at 7 lakes; 62% were western grebes, 15% were Clark's grebes, and 23% could not be identified to species. THg concentrations in grebe eggs did not differ between species ($F_{2,92.31}$ = 0.64, p = 0.53) or egg collection status ($F_{1,92.06}$ = 2.10, p = 0.15). Least squares mean THg concentrations in grebe eggs ranged from 0.03 ± 0.01 μg/g fww at Big Lake to 0.15 ± 0.02 μg/g fww at Clear Lake (see Figure S2b).

We collected 505 prey fish of 14 species from 25 lakes (predominantly bluegill [*Lepomis macrochirus*], Mississippi silverside [*Menidia audens*], and threadfin shad [*Dorosoma petenense*]; see Table S1). THg concentrations in prey fish differed among species ($F_{13,374.7}$ = 9.90, p < 0.0001) while accounting for standard length ($F_{1,455.1}$ = 2.86, p = 0.09), and there was a significant species × standard length interaction ($F_{13,459.1}$ = 11.13, p < 0.0001). Least squares mean THg concentrations in prey fish ranged from 0.03 ± 0.01 μg/g dw at Eagle Lake to 0.70 ± 0.18 μg/g dw at Bridgeport Reservoir (see Figure S2c).

We collected 230 sport fish of 5 taxa from 24 lakes (predominantly largemouth bass [*Micropterus salmoides*] and rainbow trout [*Oncorhynchus mykiss*]; see Table S2). THg concentrations in sport fish differed among species ($F_{4,27.52}$ = 9.02, p < 0.0001) and increased with total length ($F_{1,208.4}$ = 30.05, p < 0.0001), while accounting for the potential species × total length interaction ($F_{4,210.5}$ = 1.77, p = 0.14). Least squares mean THg concentrations in sport fish ranged from 0.20 ± 0.06 μg/g dw at Perris Reservoir to 2.12 ± 0.63 μg/g dw at Lake Berryessa (see Figure S2d).

Factors Influencing Mercury in Grebe Blood. The most parsimonious model describing THg concentrations in grebe blood included least squares mean THg concentrations in prey fish, grebe species, grebe sex, wing molt index, and lake perimeter (see Table S3). Fifteen other models were within ΔAICc ≤ 2.0, and all included the variables least squares mean THg concentrations in prey fish, grebe species, and grebe sex. In fact, all models containing these three variables had a cumulative Akaike weight of 0.97, indicating their importance in explaining variation in grebe blood THg concentrations. The other variables that appeared in models within ΔAICc ≤ 2.0 included date, date,[2] wing molt index,[2] grebe body condition index, lake shape index, and lake area. However, these additional variables did not improve model fit and were considered to be uninformative parameters.[25] We estimated the relative importance of individual variables and found that the data strongly supported the effects of least squares mean THg concentrations in prey fish (adjusted relative variable importance [see SI] = 14.4), species (5.8), and sex (3.5), with some support for lake perimeter (1.2). In contrast, the adjusted relative variable importance for the remaining variables were all <0.

To further determine the importance of variables in the best model, we compared the best model to the same model structure but omitted one of the variables. Using this evidence ratio approach, we estimated that the best model that included least squares mean THg concentrations in prey fish was 2.47 × 10^6 times more likely than the same model without the effect of least squares mean THg concentrations in prey fish. Similarly, the best model was 428 times more likely than the same model without grebe sex, 318 times more likely than the same model without grebe species, 5.5 times more likely than the same model without lake perimeter, and only 1.03 times more likely than the same model without wing molt index.

The conditional model-averaged coefficients indicated that each 1.0 μg/g dw (approximately 0.24 μg/g ww) increase in THg concentrations in prey fish resulted in a 103% increase in THg concentrations in grebe blood. Predicted THg concentrations in grebe blood increased by 824% from 0.26 μg/g ww to 2.37 μg/g ww over the observed range of THg concentrations in prey fish among lakes (0.03 ± 0.01 μg/g dw at Eagle Lake to 0.70 ± 0.18 μg/g dw at Bridgeport Reservoir; Figure 1a). Least squares mean THg concentrations in blood were 27% higher in Clark's grebes (0.78 ± 0.10 μg/g ww) than in western grebes (0.61 ± 0.07 μg/g ww), and 22% higher in males (0.77 ± 0.10 μg/g ww) than females (0.63 ± 0.08 μg/g ww). Lastly, conditional model-averaged coefficients indicated that THg concentrations in grebe blood increased by 0.5% with each 1 km increase in lake perimeter.

Factors Influencing Mercury in Grebe Eggs. The most parsimonious model describing THg concentrations in grebe eggs included least squares mean THg concentrations in prey fish, date, and lake perimeter (see Table S4). Fifteen other models were within ΔAICc ≤ 2.0. The other variables that appeared in models within ΔAICc ≤ 2.0 included lake area, lake shape index, egg collection type, and date,[2] but these additional variables were considered to be uninformative parameters. We estimated the relative importance of individual variables and found that the data supported only the effects of least squares mean THg concentrations in prey fish (adjusted relative variable importance = 1.9) because the adjusted relative variable importance for the remaining variables were all <0.

Using evidence ratios, we estimated that the best model, which included least squares mean THg concentrations in prey fish, was 40 times more likely than the same model without the effect of THg concentrations in prey fish. Similarly, the best model was only 1.4 times more likely than the same model without lake perimeter and 1.3 times more likely than the same model without date.

Similar to THg concentrations in grebe blood, conditional model-averaged coefficients indicated that each 1.0 μg/g dw increase in THg concentrations in prey fish resulted in a 92% increase in THg concentrations in grebe eggs. Predicted THg concentrations in grebe eggs increased by 500% from 0.04 μg/g fww to 0.24 μg/g fww over the observed range of THg concentrations in prey fish among lakes (Figure 1b).

Factors Influencing Mercury in Sport Fish. The most parsimonious model describing THg concentrations in sport fish included least squares mean THg concentrations in prey fish, sport fish species, sport fish total length, lake elevation, lake area, and a sport fish species × total length interaction (see Table S5). Five other models were within ΔAICc ≤ 2.0, and all included the variables least squares mean THg concentrations in prey fish, sport fish species, sport fish length, and a sport fish species × total length interaction. In fact, all models containing these variables had a cumulative Akaike weight of 0.89. The other variables that appeared in models within ΔAICc ≤ 2.0 included lake perimeter, lake shape, and date, but these additional variables were considered to be uninformative parameters. We estimated the relative importance of individual variables and found that the data strongly supported the effects of sport fish total length (adjusted relative variable importance >36), least squares mean THg concentrations in prey fish (8.3), lake elevation (4.1), sport fish species (2.0), and sport fish species × total length interaction (3.5), with a little support for

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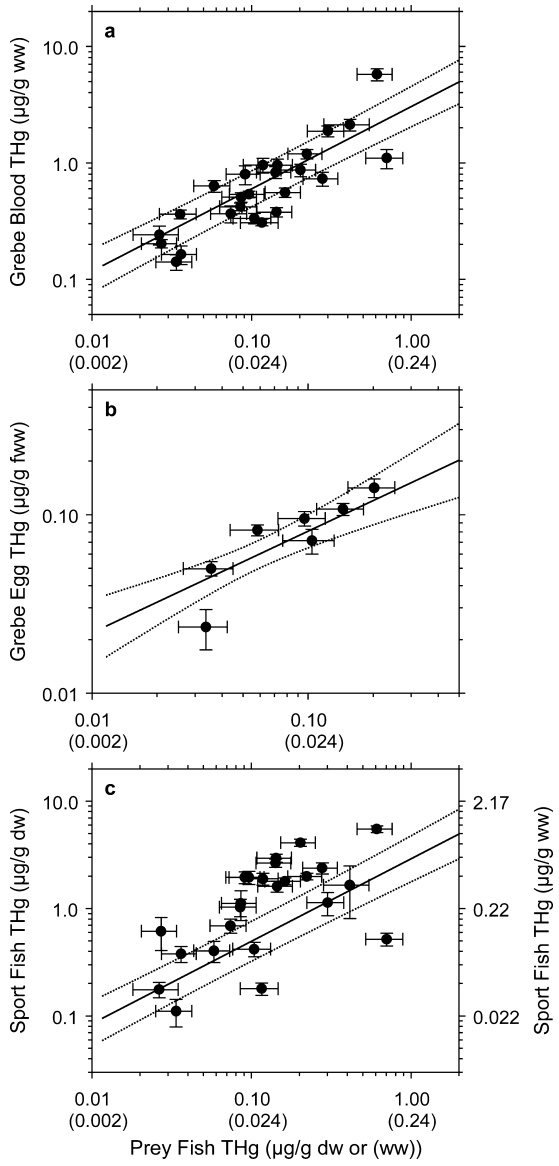

Figure 1. Total mercury concentrations in grebe blood (a; THg μg/g ww), grebe eggs (b; THg μg/g fww), and sport fish (c; THg μg/g dw [left axis] or THg μg/g ww [right axis]) versus THg concentrations in prey fish (THg μg/g dw [top row] or THg μg/g ww [bottom row]) sampled at up to 25 lakes in California, 2012−2013. *Y*-axis values are geometric means ± standard errors and *x*-axis values are least squares means ± standard errors from a global model accounting for species, standard length, and species × length interaction, with lake as a random effect. The solid line is the model-averaged predicted THg concentration and the stippled lines are the 95% confidence limits of the model-averaged predicted THg concentration. Model predictions were generated by setting all other variables in the predictive model to their mean values (or mode for wing molt and median for date), except for total length of sport fish, which was set to 350 mm. See eqs S2, S8, and S10.

lake area (0.2). In contrast, the adjusted relative variable importance for the remaining variables were all <0.

Using evidence ratios, we estimated that the best model, which included least squares mean THg concentrations in prey fish, was 1.71×10^5 times more likely than the same model without the effect of THg concentrations in prey fish. Similarly, the best model was 4.18×10^{28} times more likely than the same model without sport fish length and the sport fish species × length interaction, 49.1 times more likely than the same model without lake elevation, 44.9 times more likely than the same model without the sport fish species × length interaction, 17.3 times more likely than the same model without sport fish species and the sport fish species × length interaction, and only 1.2 times more likely than the same model without lake area.

The conditional model-averaged coefficients indicated that each 1.0 μg/g dw increase in THg concentrations in prey fish results in a 116% increase in THg concentrations in sport fish. Predicted THg concentrations in sport fish increased by 1023% from 0.20 μg/g dw to 2.21 μg/g dw over the observed range of THg concentrations in prey fish among lakes (Figure 1c). With each 10 cm increase in total length of sport fish, conditional model-averaged coefficients indicated that THg concentrations in sport fish increased by 102% for largemouth bass and 93% for rainbow trout. Lastly, conditional model-averaged coefficients indicated that THg concentrations in sport fish decreased by 28% with each 0.5 km increase in the lake's elevation.

Predictive Equations. THg concentrations in grebe blood (μg/g ww), grebe eggs (μg/g fww), and sport fish (μg/g dw) were estimated using model-averaged coefficients from our full candidate model set. See SI for all the predictive equations and model development. We compared model-averaged predictions to our individual raw THg concentrations and found good agreement for both the more complex models (grebe blood: $R^2 = 0.61$; grebe eggs: $R^2 = 0.47$; sport fish: $R^2 = 0.83$; Figure S3) as well as the simplified models that only included THg concentrations in prey fish (grebe blood: $R^2 = 0.52$; grebe eggs: $R^2 = 0.43$; but not as good for sport fish: $R^2 = 0.29$). The simplified equations to predict THg concentrations in grebe blood and eggs are as follows:

$$\ln(\mathit{FemaleWesternGrebeBlood}\,\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{ww}}) = 0.895 + 0.706(\ln(\mathit{PreyFish}\,\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{dw}})) \quad (1)$$

$$\ln(\mathit{MaleWesternGrebeBlood}\,\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{ww}}) = 1.10 + 0.706(\ln(\mathit{PreyFish}\,\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{dw}})) \quad (2)$$

$$\ln(\mathit{FemaleClark'sGrebeBlood}\,\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{ww}}) = 1.13 + 0.706(\ln(\mathit{PreyFish}\,\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{dw}})) \quad (3)$$

$$\ln(\mathit{MaleClark'sGrebeBlood}\,\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{ww}}) = 1.33 + 0.706(\ln(\mathit{PreyFish}\,\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{dw}})) \quad (4)$$

$$\ln(\mathit{GrebeBlood}\,\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{ww}}) = 1.11 + 0.706(\ln(\mathit{PreyFish}\,\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{dw}})) \quad (5)$$

$$\ln(\mathit{GrebeEgg}\,\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{fww}}) = -1.21 + 0.569(\ln(\mathit{PreyFish}\,\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{dw}})) \quad (6)$$

where $\mathit{PreyFish}\,\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{dw}}$ is the least squares mean THg concentration in prey fish at a lake.

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Mercury Correlations between Grebe Blood, Grebe Eggs, and Sport Fish. Least squares mean THg concentrations were correlated among tissues; grebe blood was related to sport fish (n = 24 lakes, R^2 = 0.59, $F_{1,22}$ = 31.79, p < 0.0001; Figure 2a top panel), grebe eggs were related to sport fish (n = 6 lakes, R^2 = 0.67, $F_{1,4}$ = 8.17, p = 0.05; Figure 2b), and grebe eggs were strongly related to grebe blood (n = 7 lakes, R^2 = 0.93, $F_{1,5}$ = 71.43, p < 0.001; Figure 2c). Equations for these relationships are as follows:

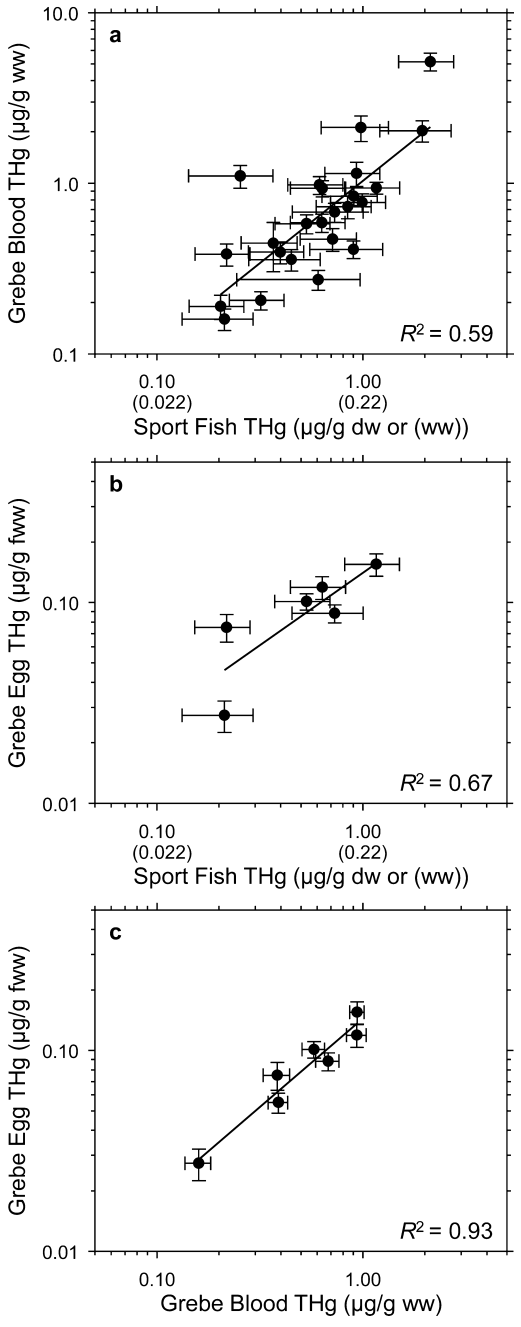

Figure 2. Total mercury concentrations (THg) in (a) grebe blood versus sport fish, (b) grebe eggs versus sport fish, and (c) grebe eggs versus grebe blood at up to 25 lakes in California, 2012–2013. Values are least squares means ± standard errors from separate models for each tissue. X-axis values for THg concentrations in sport fish (a, b) are displayed on a dry weight (THg µg/g dw [top row]) and wet weight (THg µg/g ww [bottom row]) basis. See eqs 7-9 in Results.

$$\ln(GrebeBlood\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{ww}}) = 0.03 + 0.966(\ln(SportFish\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{dw}})) \quad (7)$$

$$\ln(GrebeEgg\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{fww}}) = -1.97 + 0.720(\ln(SportFish\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{dw}})) \quad (8)$$

$$\ln(GrebeEgg\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{fww}}) = -1.94 + 0.883(\ln(GrebeBlood\overline{\mathrm{THg}}_{\frac{\mu g}{g}_{ww}})) \quad (9)$$

Literature Review. We reviewed the literature for available relationships between THg concentrations in birds and prey fish, and found six studies with such correlations.[13,15,16,26−28] We then extracted each study's equation and compared it to our equations for female western grebes and Clark's grebes (Figure 3). We were unable to determine the equation and bird's tissue type used in Hosseini et al.;[28] we therefore excluded that study from further consideration. The remaining studies all focused on the relationship between THg concentrations in common loons and their fish prey. It was necessary to make several transformations to the equations in order to make them comparable across studies. We extracted the equations specific to female common loons when possible,[13,15,16,26,27] but the equation was not sex-specific for adult loons in the study by Scheuhammer et al.[16] We included both equations provided by Evers et al.[13] that were specific to two different size classes of prey fish (i.e., 50−100 mm and 100−150 mm). Champoux et al.[26] did not present an equation; we therefore extracted their equation using the provided slope

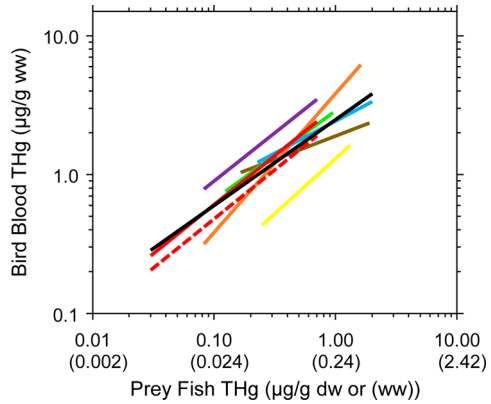

Figure 3. Relationships between total mercury concentrations in bird blood (THg µg/g ww) and THg concentrations in prey fish (THg µg/g dw [top row] or THg µg/g ww [bottom row]) produced by six studies on either common loons (purple line [50−100 mm prey fish],[13] green line [100−150 mm prey fish],[13] blue line,[16] yellow line,[26] orange line,[15] brown line[27]), western grebes (stippled red line[this study]), or Clark's grebes (solid red line[this study]). Regressions are specific to female birds, except for the study by Scheuhammer et al.[16] which included both males and females. We extracted each study's equation and plotted it over the reported range of THg concentrations in prey fish specific to each study. The solid black line is the average relationship between THg concentrations in bird blood and THg concentrations in prey fish (see eqs 10 and 11 in Results), which was calculated by averaging the data across studies where they overlapped in THg concentrations in prey fish.

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in the Spearman correlation and estimating the intercept graphically from their figure. Finally, the equation provided by Yu et al.[27] included an additional variable for MeHg concentrations in zooplankton. We simplified this equation by using their average reported MeHg concentration in zooplankton (0.07 μg/g dw). After extracting these equations, we plotted each equation over the reported range of THg concentrations in prey fish specific to each study, and, when necessary, converted THg concentrations in prey fish from wet weight to dry weight using an average prey fish moisture content of 75.8% (Figure 3). Lastly, we developed an average relationship between THg concentrations in bird blood and THg concentrations in prey fish (on both a ww and dw basis) by averaging the data across studies wherever they overlapped in THg concentrations in prey fish (Figure 3):

$$\ln(FemaleBirdBlood\overline{THg}_{\frac{\mu g}{g}ww}) = 1.788 + 0.6182(\ln(PreyFish\overline{THg}_{\frac{\mu g}{g}ww})) \quad (10)$$

$$\ln(FemaleBirdBlood\overline{THg}_{\frac{\mu g}{g}ww}) = 0.9114 + 0.6182(\ln(PreyFish\overline{THg}_{\frac{\mu g}{g}dw})) \quad (11)$$

We evaluated the goodness of fit of the general model (eq 11) by comparing the predicted THg concentration obtained with the actual least squares mean THg concentrations in grebe blood at each lake and found that the fit was strong ($R^2 = 0.73$).

DISCUSSION

THg concentrations in piscivorous birds, represented by western grebes and Clark's grebes, were strongly correlated with THg concentrations in prey fish among lakes in California (Figure 1a,b). Similarly, THg concentrations in prey fish also were a strong predictor of THg concentrations in sport fish (Figure 1c). Observed THg concentrations among lakes ranged by more than 32-fold in grebe blood, 10-fold in sport fish, and 23-fold in prey fish, and provided the necessary data to apply our model over a wide range of environmentally relevant Hg concentrations (Figure S2). Using a model-averaging approach, we developed equations to predict THg concentrations in bird blood (eqs S1−S7), bird eggs (eq S8), and sport fish (eqs S10−S12) using THg concentrations in prey fish, sampling date, lake attributes, and animal-specific variables (such as species, sex, and body condition of grebes). Because THg concentrations in prey fish were so strongly correlated with THg concentrations in piscivorous birds, we also were able to develop more simplistic, but still reliable, models with just THg concentrations in prey fish (eqs 1−6). Caution should be used when extending these equations past the observed data. Each 1.0 μg/g dw (approximately 0.24 μg/g ww) increase in THg concentrations in prey fish resulted in an increase in THg concentrations of 103% in grebe blood, 92% in grebe eggs, and 116% in sport fish. We also found strong correlations between THg concentrations in birds and sport fish among lakes (Figure 2; eqs 7−9), although the relationships with THg concentrations in prey fish were generally stronger (Figure 1).

There are several studies in North America that have demonstrated strong correlations between THg concentrations in common loons and THg concentrations in small prey fish,[13,15,16,26,27] but there are few other studies assessing the relationship between Hg concentrations in fish and other bird species besides loons.[28] We extracted each of these five studies' equations predicting THg concentrations in blood of common loons from THg concentrations in prey and compared them to our equations for female western grebes and Clark's grebes (Figure 3). Interestingly, most of these equations yielded similar predicted THg concentrations in bird blood based on THg concentrations in prey fish. To illustrate, we can use the derived benchmark of 0.1 μg/g ww in prey fish for adverse behavioral impacts to adult common loons.[29] Using this value of 0.1 μg/g ww in prey fish (approximately 0.41 μg/g dw using 75.8% moisture content), female western grebes and Clark's grebes foraging on these fish would be predicted to have a THg concentration of 1.31 μg/g ww and 1.66 μg/g ww in their blood, respectively. In comparison, 0.1 μg/g ww in prey fish would be predicted to result in THg concentrations in female common loon blood of 1.34 μg/g ww,[27] 1.56 μg/g ww[16] (using an average moisture content of 75.8%), 1.57 μg/g ww (using the equation for 100−150 mm prey fish),[13] and 1.58 μg/g ww.[15] However, 0.1 μg/g ww in prey fish would be predicted to result in a lower THg concentration in blood of female common loons (approximately 0.65 μg/g ww) in the study by Champoux et al.[26] In general, these results indicate that it is feasible to estimate Hg exposure risk of piscivorous birds from THg concentrations in prey fish.

In addition to THg concentrations in prey fish, bird species and sex influenced THg concentrations in grebe blood. THg concentrations in blood were 27% higher in Clark's grebes than in western grebes, and 22% higher in males than in females. Higher THg concentrations in male than female birds is common,[15,16,30−32] but the mechanism is unclear. It is possible that male grebes had higher THg concentrations than females because they were larger (19% heavier on average) and might consume more or larger prey. However, potential differences in diet between the sexes are not well studied.[33,34] This explanation has been postulated as the reason for sex differences in THg concentrations of common loons,[16,32,35] although sexual size dimorphism does not occur in some other bird species where higher THg concentrations in males than females occurs.[30,31] Additionally, size differences do not appear to explain the higher THg concentrations in Clark's grebes than in western grebes, because body mass (3% smaller) and culmen length (3% smaller) were both slightly smaller in Clark's grebes than in western grebes.

Generally, lake attributes appeared to play a small role in THg concentrations in grebes and sport fish. There was some limited support for an influence of lake perimeter on THg concentrations in grebe blood; however, the effect size was relatively small with THg concentrations in grebe blood estimated to have increased by only 0.5% with each 1 km increase in lake perimeter. For sport fish, there was stronger support for an influence of lake elevation on THg concentrations, with THg concentrations in sport fish decreasing by 28% with each 0.5 km increase in lake elevation. There are several potential mechanisms that could lead to THg concentrations in birds and fish to be correlated with general lake attributes, and lakes with larger perimeters and at lower elevations may have biogeochemical characteristics that enhance MeHg production.[36,37] Despite these potential effects, THg concentrations in prey fish were the most important factor influencing Hg concentrations in birds.

Monitoring programs assessing Hg contamination of aquatic ecosystems typically sample fish rather than wildlife due to the relative ease of sampling fish and their more direct link to

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human exposure.[5−7] This often means that the risk of Hg exposure to aquatic wildlife is not known in many Hg monitoring programs. We developed predictive equations to link THg concentrations in fish to those in piscivorous wildlife so that Hg monitoring programs that sample fish, and do not directly sample wildlife, can more easily estimate risk of Hg exposure to birds. Using these equations, grebes would be predicted to have a moderate risk THg concentration of 1.6 μg/g ww in their blood when feeding on prey fish at the THg benchmark of 0.1 μg/g ww.[29] Although differences among bird species, such as prey selection and bioenergetics, and lake-specific biogeochemistry could result in substantially different MeHg biomagnification rates for similar THg concentrations in prey fish, we nonetheless found that most of the available predictive equations have resulted in similar THg concentrations in bird blood (Figure 3). Predictive equations specific to the species of bird and area of study are preferred (such as eqs 1−6), but the general equation we developed in this study (eqs 10 and 11) could be used to approximate Hg exposure of piscivorous birds when no other reasonable estimate is available.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b02691.

Map of study lakes (Figure S1), THg concentrations among lakes (Figure S2), predictive equations (eqs S1−S15), predictive model fit (Figure S3), number and species of fish sampled (Tables S1 and S2), AIC tables (Tables S3−S5), grebe sampling methods, THg determination and quality assurance methods, and additional statistical methods are reported in SI (PDF)

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Notes

The authors declare no competing financial interest.

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